International Journal of Thermal Sciences 129 (2018) 301–308

Contents lists available at ScienceDirect

International Journal of Thermal Sciences

journal homepage: www.elsevier.com/locate/ijts

Measurement of radial thermal conductivity of a cylinder using a time-varying heat flux method

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ARTICLE INFO

Keywords:
Thermal conductivity measurement
Radial thermal conductivity
Heat transfer modeling
Li-ion cell

ABSTRACT

Despite a variety of techniques available for thermal conductivity measurement, there is a lack of methods to directly measure the radial thermal conductivity of a cylinder. This presents a critical gap in heat transfer metrology, particularly in presence of anisotropic thermal conduction, such as in a Li-ion cell, where radial thermal conductivity plays a key role in determining performance and safety. This paper reports a technique for measurement of radial thermal conductivity of a cylinder by accounting for variable heat flux into the cylinder when heated on the outside. It is shown that heat flux into the cylinder from a thin heater wrapped around its surface varies significantly with time. This variation, which was neglected in past work, is accounted for by developing a variable heat flux model for experimental data analysis. For two different materials, measurements of radial thermal conductivity using this approach are shown to be in close agreement with standard thermal conductivity measurement using the transient plane source method. Radial thermal conductivity of a 26650 Li-ion cell is measured to be 0.39 W/mK. Besides contributing a new approach for thermal metrology in cylindrical systems, this work also improves the understanding of thermal phenomena in Li-ion cells.

1. Introduction

Thermal conductivity and heat capacity are the two key thermophysical properties that determine the thermal performance of any material, component or system [1,2]. Thermal conductivity of a body is defined based on Fourier law of thermal conduction [2,3]. For most engineering materials, thermal conductivity is isotropic, although in some cases, such as in a Li-ion cell, the value of the thermal conductivity may be strongly direction-dependent [4,5]. A number of experimental methods have been used for measurement of thermal properties of engineering materials, components and systems [4,6–8]. Most of these methods compare experimental measurement of the thermal response to an imposed heat flux with an appropriate analytical model to determine thermal conductivity, and in some cases, heat capacity as well. For example, several methods impose a one-dimensional heat flux through the body of interest and measure temperature difference across the body to determine the thermal conductivity [9]. The effect of thermal contact resistance in such measurements has been accounted for [10]. Laser flash methods impose a heat flux on one face of the test sample and measure the transient thermal response on the other face in order to determine thermal conductivity [6]. Comparison of the short-time thermal response to transient heating of a test sample with an analytical model for thermal conduction in an infinite medium has also been used to measure thermal conductivity [7]. Response to steady-periodic heating has also been used to determine thermal properties of materials [11]. Many of these methods require specific geometries for test samples and are suited for measurement in only specific directions. In particular, several of these methods are inappropriate for measurement of radial thermal conductivity of cylindrical samples. This may not be an important concern for bodies with known isotropy in thermal conductivity, wherein the measurement can be easily carried out in the axial direction instead. However, some cylindrical bodies exhibit anisotropy, making it necessary to directly measure the radial component of thermal conductivity. One-dimensional heat flux methods are not possible to use in such a case, since imposition of a heat flux on the outer radial surface of a cylinder does not produce a steady-state [4], and since measurement of temperature inside a cylinder is often not possible [12–14]. Plane heat source based methods are also not appropriate since a heat source on the outer surface of the cell may result in heat conduction in all three directions, making it impossible to isolate only the radial component. Laser flash based methods are also not appropriate, since these methods require a sample with flat faces for both heat pulse absorption and temperature measurement. Development of methods to measure the radial thermal conductivity is therefore of much interest.

Measurement of radial thermal conductivity is important for

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https://doi.org/10.1016/j.ijthermalsci.2018.03.008
Received 15 December 2017; Received in revised form 6 February 2018; Accepted 10 March 2018
1290-0729/

understanding the thermal safety of cylindrically shaped engineering systems such as a Li-ion cell. Due to heat generation during charging and discharging processes [15], there is significant temperature rise in a Li-ion cell [15–17], which leads to serious safety and performance challenges. Particularly, temperature rise beyond a certain threshold initiates multiple, cascaded exothermic reactions that result in an undesirable thermal runaway scenario involving uncontrolled temperature rise and catastrophic failure [18–20]. Radial thermal conductivity of a cylindrical Li-ion cell is a key thermal property, the value of which is critical for accurate pre-operation design, as well as run-time thermal performance modeling and prediction. Unlike heat capacity, which can be easily measured through calorimetry-based techniques, only a limited amount of literature exists on measurement of this important thermal property [4,5]. Drake et al. [4] have reported radial thermal conductivity measurement by heating the cell on its outer radial surface with a thin heater and measuring the resulting temperature rise as a function of time. Measurements indicated very strong anisotropy, highlighted by a 150X difference in radial and axial thermal conductivities. This measurement was based on an analytical model that assumed constant heat flux into the cell as a function of time. However, it is possible that heat flux may actually vary with time due to heat conduction into the insulation material, and due to heating up of the thin heater material itself. The first effect, while not important for a steady-state measurement, may be significant for a transient measurement such as the one conducted by Drake et al. The second effect can be minimized by reducing the thermal mass of the heater, but cannot be eliminated completely. As a result of these effects, it is necessary to redefine the thermal conductivity measurement methodology by either modifying the analytical model to account for heat losses, or by modifying the experimental conditions in order to minimize or compensate for such heat losses. Such corrections may increase the accuracy of radial thermal conductivity measurements using this method.

This paper presents experimental measurements to demonstrate the presence of significant variation in heat flux as a function of time in experiments for radial thermal conductivity measurements. Further, an approach to correct for this effect is demonstrated through experiments and data analysis. A modified analytical model capable of accounting for time-varying heat flux into the cylinder is presented. Heat flux measurements are combined with optimization of experimental time duration through sensitivity analysis to accurately determine the radial thermal conductivity. It is shown that this approach results in accurate measurement of radial thermal conductivity of two test materials, which are found in both cases to be in good agreement with measurements based on a separate, independent method. This variable heat flux based approach is implemented for measurement of radial thermal conductivity of a 26650 Li-ion cell. Analysis of this method shows significant dependence of radial thermal conductivity on the assumed value of heat capacity.

2. Mathematical model

2.1. Variable heat flux model

Consider a cylindrical body of radius R subjected to a certain time-varying heat flux on its outer surface at $r = R$ due to heat generated in a thin heater wrapped around its outer radial surface. The interest is in predicting temperature rise on the outer surface of the cylinder as a function of time, which may be compared against experimental measurements to determine the thermal properties of the cylinder. Specifically, measurement of the radial thermal conductivity is of interest, since the heat capacity can be easily measured using calorimetry-based techniques. Assuming the radial thermal conductivity, heat capacity and density of the body to be k_r, C_p and ρ respectively, the energy conservation equation governing the temperature rise $\theta(r,t)$ in the cylinder is given by

$$\frac{k_r}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\theta}{\partial r}\right) = \rho C_p\frac{\partial\theta}{\partial t} \tag{1}$$

where r and t are the radial and time coordinates respectively.

Since the cell is being heated on the outer surface, a heat flux boundary condition applies at $r = R$. Drake et al. solved this problem for constant heat flux on the outer surface [4]. However, as shown by experimental data discussed in section 4.1, heat flux into the test samples does not remain constant with time, even if heat is generated at a constant rate in the heater. As a result, it is important to re-derive a solution for equation (1) to account for time-varying heat flux into the cylinder on the outer surface, $Q(t)$, instead of assuming constant heat flux. This complicates the problem somewhat due to a time-varying boundary condition given by

$$\frac{\partial\theta}{\partial r} = \frac{Q(t)}{k_r} \quad \text{at } r = R \tag{2}$$

An additional boundary condition is that

$$\frac{\partial\theta}{\partial r} = 0 \quad \text{at } r = 0 \tag{3}$$

Finally, temperature rise is assumed to be zero at the initial time. Note that in general, the temperature distribution $\theta(r,t)$ may not reach a steady state unless $Q(t)$ becomes zero at large times, which is not the case here, since the heater around the body continues to generate heat. This lack of a steady state is similar to the problem solved by Drake et al. [4], and can be addressed by subtracting from $\theta(r,t)$ the average temperature rise as a function of time, which in this case is given by $\theta_{mean}(t) = \frac{2\cdot\int_0^t Q(\tau)d\tau}{\rho C_p R}$.

Once this transformation is carried out, the remaining problem still involves a time-varying heat flux at the outer surface, but is nevertheless easier to solve as it does have a steady-state. This problem is solved using the method of undetermined parameters by assuming a series solution comprising time-dependent coefficients $c_n(t)$ and eigenfunctions of the corresponding homogeneous problem [21].

$$\theta(r,t) - \theta_{mean}(t) = \sum_{n=1}^{\infty} c_n(t)J_0(\lambda_n r) \tag{4}$$

Where J_0 denotes Bessel function of the first kind of order 0 and the eigenvalues λ_n are obtained from the roots of J_1, the Bessel function of the first kind and of order 1. By inserting the assumed form of the solution, equation (4) into the governing energy equation and using the boundary conditions to simplify, the following ordinary differential equation can be derived for $c_n(t)$

$$c'_n(t) + \alpha_r\lambda_n^2 c_n(t) = \frac{\alpha_r R J_0(\lambda_n R)}{N_n\cdot k_r}Q(t) \tag{5}$$

where N_n are norms of the eigenfunctions [2] and $\alpha_r = {}^{k}\!/_{\rho C_p}$ is the radial thermal diffusivity.

Equation (5) can be solved along with the zero initial condition for $c_n(t)$ to result in the following final form of the solution for the temperature rise.

$$\theta(r,t) = \frac{2}{\rho C_p R}\int_0^t Q(\tau)d\tau + \sum_{n=1}^{\infty}\frac{\alpha_r R J_0(\lambda_n R)}{N_n k_r}\int_0^t Q(\tau)\exp(-\alpha_r{\lambda_n}^2(t-\tau))d\tau \tag{6}$$

Equation (6) represents the analytically derived temperature rise as a function of time that can be compared with experimental data to determine the radial thermal conductivity. This requires measurement of heat flux $Q(t)$ and evaluation of integrals involving $Q(t)$ in equation (6).

For a special case where $Q(t) = Q_0$ is constant, such as the case considered by Drake et al. [4], equation (6) can be simplified to